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MEASUREMENTS OF THE AGE OF THE SOLAR SYSTEM[1]

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For over a century astronomers have sought a satisfactory explanation for the origin of the earth and the solar system. Successive hypotheses have had some transient successes, but in the end additional astrophysical data or more penetrating mathematical analyses have shown all the hypotheses to be unsound. Recently the hypothesis that the solar system was formed by the condensation of matter produced by a near encounter between two stars has also been shown to be untenable. Spitzer[2] has noted that a filament of stellar material produced by an encounter between two stars would have a temperature in excess of a million degrees centigrade, and he has shown that the gases within such a filament would accelerate outwards and within a few hours would reach the velocity of escape. Thus it seems very doubtful that the stellar encounter theory can survive as an explanation of the origin of the solar system.

As an aid to the construction at some later date of a satisfactory theory concerning the origin of the solar system, it is important to make as many accurate measurements as possible on the physical constants of the solar system. Such measurements include the age of the earth and the age of as many non-terrestrial samples of the solar system as it is possible to obtain.

The only non-terrestrial samples that can be brought to the laboratory are recovered meteorites. A meteorite having a heliocentric velocity, at the earth's distance from the sun, greater than

[1] From a paper presented at the Interamerican Astrophysical Congress at Puebla, Mexico, February 25, 1942.

[2] Lyman Spitzer, Jr., The Astrophysical Journal, **90**, pp. 675–788, 1939.

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42.4 km. per second can escape from the sun's gravitational field. Presumably such a meteorite would represent a sample of extra-solar astronomical material. Meteorites having a heliocentric velocity less than 42.4 km. per second are therefore to be taken as extra-terrestrial samples of the solar system.

It is known that distinctly more meteorites strike the surface of the earth between the hours of noon and midnight than in the morning hours between midnight and noon. The direction of the earth's revolution around the sun is such that the forward surface of the earth, which sweeps the space through which the earth travels, is always the half whose time is between midnight and noon. Extra-solar meteorites could be expected to show no preferred direction in space, and should appear randomly in the zone swept out by the earth's motion around the sun. Consequently a predominance of morning falls should be observed if all meteorites were of extra-solar origin. The fact that the majority of the falls are in the afternoon shows that the majority of meteorites move in "direct" orbits, whose direction around the sun is the same as that of the earth's. Paneth[1] ably extends this argument and concludes that all meteorites have since their formation been members of the solar system. Several very bright meteors have been photographed by Whipple,[2] who used two cameras, equipped with "light-chopper" shutters, at the ends of a 24-mile base line near Cambridge, Massachusetts. The heliocentric velocities determined from these observations show that the bright meteors observed so far are of solar origin. However, none of these meteors was sufficiently large to allow it to penetrate the earth's atmosphere completely and land on the earth's surface as a meteorite.

In performing age measurements on meteorite specimens it is important to deal, if possible, with material known to be either of solar or extra-solar origin. Unfortunately one of the rarest astronomical events is the accurate observation of the velocity of a meteor, followed by the actual recovery of the meteorite, if any, resulting from the fall. Circumstances are seldom so favorable. In the painstaking catalogue of *611 Orbits of Big Meteors*, edited by Hoffmeister in 1926 from the life work of von Niessl, only seven recovered meteorites could be named for which any kind of velocity observations had been made while the parent meteor fell through

[1] F. A. Paneth, "The Origin of Meteorites." The Haley Lecture, published by the Clarendon Press, Oxford, 1940.

[2] F. L. Whipple, Proc. Amer. Phil. Soc., 79, p. 449, 1939.

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the earth's atmosphere. Of the seven the velocity data on the Rochester and St. Michel meteorites were too doubtful to deserve serious consideration. The available data on the remaining five meteors led to computed heliocentric velocities which follow: Pultusk, 56 km. per second; Homestead, 40; Krähenberg, 57; Orgueil, 52; Treysa, 38.

The largest amount of velocity data available for any meteorite relates to the great fall of stones in Pultusk, Poland, in January, 1868. T. G. Galle[1] interviewed many people who observed this meteorite fall and concluded from these data that the heliocentric velocity of the Pultusk meteorite was definitely greater than the velocity of escape from the solar system. These original observations have recently been reviewed by Professor C. C. Wylie,[2] whose extensive experience in such interviews leads him to reduce the reported height of appearance of the meteorite, from Galle's estimate of over 100 miles to 52 miles, which corresponds to the measured height of appearance of modern spectacular meteors. This has the effect of reducing the computed velocity so that the Pultusk meteorite may have originated in the solar system and would have had an elliptic orbit resembling those of the minor asteroids. We thus assume that the Pultusk meteorite, on which extensive age measurements have been made by methods based on radioactivity, originated in the solar system.

HELIUM AGE MEASUREMENTS

It is well known that the decay of long-lived radioactive substances such as uranium offers a general method of age determination. It has been proved that the rate of radioactive decay does not depend on the age of the atoms involved, and consequently that the decay of uranium and thorium into their daughter products has always proceeded at a uniform rate. Helium is one of the products of these radioactive disintegrations. Assuming that all of the helium formed has remained trapped in the specimen, the age of any sample of terrestrial or meteoritic material is proportional to the ratio of the total amount of helium produced in the sample to the amount of uranium and thorium present. This method of age analysis has been used extensively in the study of terrestrial materials.[3]

[1] T. G. Galle's original papers have been translated and republished serially by W. H. Hass, Popular Astronomy, October, 1942, et seq.

[2] C. C. Wylie, Science, 9, p. 264, 1939.

[3] For example: C. Goodman and R. D. Evans, Bull. Geol. Soc. Amer., 52, pp. 491–544, 1941; C. Goodman, Jour. App. Phys., 13, p. 276, 1942.

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The helium method was first developed and applied to meteorites by F. A. Paneth and his students. In their original studies[1] of twenty-three iron meteorites these investigators showed that helium does not leak out of iron meteorites at ordinary temperature. Helium and radium measurements were made on all of these specimens, but it was not possible at that time to perform thorium analyses. By assuming that all of the helium was formed by the decay of uranium it was then possible to calculate maximum values for the age of these iron meteorites. This "age" corresponds to the time which has elapsed since the helium was last quantitatively expelled, or roughly the time which has elapsed since the material last cooled below 1000° C.

The results obtained by Paneth and his co-workers are presented graphically in figure 30. It will be noted that there is an essentially uniform distribution of ages from a maximum of 2,800 million years down to essentially zero (i.e. 100 million years), with no tendency toward grouping about any favored time. A similar plot of the available age measurements on terrestrial surface rocks and minerals has the same general appearance.[2] This fact might at first suggest that meteorites were fragments from large astronomical bodies which broke up at various times in the past, and were hot enough to expel all their helium at the time of fragmentation. However, the well-known Widmanstätten figures, which can be simulated in the laboratory only by the very slow cooling of iron-nickel alloy, are preserved in these samples. The "age" data refer to the elapsed time since the last intense heating, that is, to the time when the existing Widmanstätten figures were formed, since they are probably destroyed by temperatures much in excess of 1000° C. The ages therefore probably refer to the date of formation of a sufficiently large astronomical body to provide for very slow internal cooling. The observed distribution of ages therefore suggests that the formation of new astronomical bodies is a continuous process, possibly still proceeding at a relatively uniform rate.

In 1932 Paneth measured the helium and radium content of a specimen of the Pultusk stone.[3] This measurement led to an age of about 500 million years, but is unquestionably low by an entirely

[1] Paneth, Urry, and Koeck, Nature, March 29, 1930; Paneth, Naturwiss., 19, p. 164, 1931; Paneth and Koeck, Zeit. Phys. Chem., Bodenstein-Festband, p. 145, 1931.

[2] C. Goodman, Jour. App. Phys., 13, p. 276, 1942.

[3] Personal communication from Professor Paneth to Dr. H. Shapley.

unknown amount, because of the ease with which helium was demonstrated to escape from the porous stony mass, in contrast to its proved retention by compact iron meteorites.

More recently Professor Paneth has succeeded in carrying out thorium measurements, as well as helium and radium analyses, on

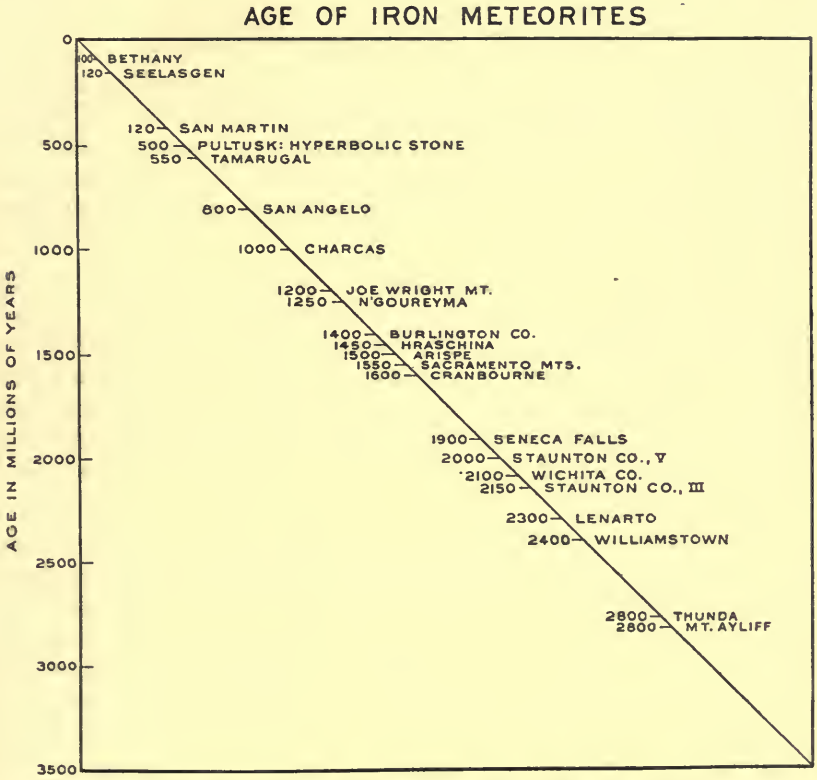

FIG. 30. Age distribution of iron meteorites, based on the helium-radium ratio. Originally these were thought to be maximum values, because of the absence of thorium measurements.[1] Basing his conclusions on new measurements of the thorium, radium and helium content of six iron meteorites,[2] Paneth now believes the radium measurements of the early series to have been too high, so that the true ages-since-solidification may have been three times as great as the values shown. This revision does not alter the conclusions given later in the present paper except by suggesting that some iron meteorites may have solidified long before the earth's crust solidified, and would therefore have helium ages greater than the age of the earth.

[1] Paneth, Urry, and Koeck, Nature, March 29, 1930; Paneth, Naturwiss., 19, p. 164, 1931; Paneth and Koeck, Zeit. Phys. Chem., Bodenstein-Festband, p. 145, 1931.

[2] Arrol, Jacobi, and Paneth, Nature, 149, pp. 235–238, 1942.

samples of four iron meteorites.[1] While the absolute accuracy of the figures is still doubtful the relative values can be taken as a significant guide. These results, in millions of years since solidification, are: Bethany, Goamus 30; San Martin 500; Bethany, Amalia 1000; Thunda >3000. Again a broad and continuous distribution in ages is exhibited.

DIFFERENTIAL AGES BY RADIOACTIVITY

The helium method is not applicable to stony meteorites, because of the inability of the stony mass to retain quantitatively the helium generated in it. Neither is it possible to make age measurements by the lead method, because of the very small amount of uranium and thorium contained in meteorites, and consequently the negligible quantity of lead generated.

Useful information can be gained, however, from a study of the relative abundance of the isotopes of radioactive elements in the meteorites. Such studies possess the unique advantage of eliminating all questions relating to the leakage or leaching of radioactive decay products such as helium or lead. Any physical or chemical processes, such as leakage or leaching, should affect all of the isotopes of the element involved in substantially the same way, leaving their ratios unaltered.

It is well known that all terrestrial specimens of any particular element always exhibit the same isotopic abundance, independent of the place of origin of the parent mineral or rock containing the element. It is also found from isotopic abundance or chemical atomic weight studies in meteorites that the isotopic abundance is the same as in terrestrial materials for all the elements studied so far,[2] namely: carbon, oxygen, silicon, chlorine, iron, cobalt, and nickel.

We shall see below that there is good justification for making the following basic assumption: that the initial isotopic ratio for any element is a constant of nature and is independent of the time or place of origin of these atoms in the galactic system.

Uranium is a mixture of independent isotopes, uranium I and actinouranium, having widely different half-periods. Thus, one-

[1] F. A. Paneth, "The Origin of Meteorites." The Haley Lecture, published by the Clarendon Press, Oxford, 1940.

[2] R. D. Evans, Popular Astronomy, **46**, pp. 159–170, 1938; A. O. Nier and A. Gulbransen, Jour. Amer. Chem. Soc., **61**, p. 697, 1939; G. E. Valley and H. H. Anderson, Phys. Rev., **59**, 113A, 1941.

half of any given initial quantity of the isotope known as uranium I will have decayed in 4,530 million years, while the half-period of the isotope actinouranium is only 710 million years. Thus, in any sample of uranium, the passage of time causes the shorter-lived isotope, actinouranium, to become progressively less abundant as compared with the long-lived isotope, uranium I.

The element potassium is a mixture of three isotopes, two of which are stable, while the third is radioactive with a half-period of 1,600 million years. Thus the radioactivity per gram of potassium, that is, the "specific radioactivity" of any sample of potassium, decreases with time, due to the decay of the one radioactive isotope.

Mass spectroscopic studies[1] of the relative abundance of the uranium isotopes in three uranium minerals from widely different localities have shown a constant abundance ratio of 139 atoms of uranium I per atom of actinouranium. Because these two isotopes decay with different radioactive periods the relative abundance changes with time. In order to explain the constant isotopic abundance found in all samples of uranium either of two assumptions can be made. It is simpler to assume that all uranium atoms were created with the same original isotopic abundance ratio, and at the same time in the past.

The only other assumption that can satisfy contemporary observations on the isotopic abundance is the very *ad hoc* and awkward assumption that after the first sample of uranium was created all successive samples were created with a relative abundance ratio so delicately altered that they would exactly match the alteration which time had imposed upon the relative abundance of the isotopes in the first sample of uranium ever created. This cumbersome assumption seems even more untenable when we consider other long-lived radioactive substances such as potassium. There is no appreciable difference between the specific radioactivity of terrestrial potassium from a wide variety of mineral sources.[2] Here a similar alteration in the isotopic abundance ratio of newly created potassium isotopes would have to be invoked, and the time factor would have to be a different one from that applied by nature to the uranium case, because the half period of the radioactive isotope of potassium is markedly different from the half periods of uranium I and actinouranium. Thus it seems much more satisfactory to assume

[1] A. O. Nier, Phys. Rev., 55, pp. 150–153, 1939.

[2] Biltz and Marcus, Z. anorg. Chem., 81, p. 369, 1913.

that the initial isotopic abundance ratio for any element is a constant of nature.

If a meteorite sample were found to have the radioactive isotopes of uranium or potassium in a relative abundance which differed from the normal terrestrial abundance, it could be inferred that the atoms of the meteorite were created at a different time than the atoms of the same element in the earth. Thus the study of the relative abundance of the isotopes of radioactive substances offers a method of determining differential ages.[1]

THE DIFFERENTIAL AGE EQUATIONS

We derive the equations relating isotopic abundances to the age of the atoms in the following manner: In any particular sample of uranium, let:

A_0 = number of atoms of actinouranium, t years ago
A = number of atoms of actinouranium at present
λ_A = radioactive decay constant of actinouranium
U_0 = number of atoms of uranium I, t years ago
U = number of atoms of uranium I at present
λ_U = radioactive decay constant of uranium I

Then the fundamental equations of radioactive decay become:

$$A = A_0 \epsilon^{-\lambda_A t} \tag{1}$$

$$U = U_0 \epsilon^{-\lambda_U t} \tag{2}$$

Dividing, and denoting a sample of meteoritic uranium by subscripts m, and a sample of terrestrial uranium by subscripts e, we have:

$$(A/U)_m = (A_0/U_0)_m \epsilon^{-(\lambda_A - \lambda_U) t_m} \tag{3}$$

$$(A/U)_e = (A_0/U_0)_e \epsilon^{-(\lambda_A - \lambda_U) t_e} \tag{4}$$

Now, dividing equation 3 by equation 4, we obtain:

$$\frac{(A/U)_m}{(A/U)_e} \frac{(A_0/U_0)_e}{(A_0/U_0)_m} = \epsilon^{-(\lambda_A - \lambda_U)(t_m - t_e)} \tag{5}$$

[1] R. D. Evans, Popular Astronomy, 46, pp. 159–170, 1938.

We now introduce our single assumption: that the initial isotopic abundance ratio was the same for all samples of uranium, regardless of when and where created; that is:

$$\frac{(A_0/U_0)_e}{(A_0/U_0)_m}=1 \tag{6}$$

Substituting equation 6 into equation 5, and taking natural logarithms of both sides, we have:

$$ln\ [(A/U)_e/(A/U)_m] = (\lambda_A-\lambda_U)\ (t_m-t_e) \tag{7}$$

where (t_m-t_e) is the difference in the time elapsed since the creation of the uranium atoms in the meteorite specimen and in a terrestrial specimen.

Meteorites and terrestrial rocks contain only the order of 10^{-6} grams of uranium per gram of sample. Consequently it is impossible to obtain enough uranium to measure the isotopic abundance ratios $(A/U)_m$ and $(A/U)_e$ by means of the mass spectrograph. We must turn to radioactive methods for the determination of these isotopic ratios.

We can reduce the argument of the logarithm in equation 7 to quantities which can be measured by radioactivity by multiplying its numerator and denominator by λ_A/λ_U, the ratio of the decay constants of actinouranium and uranium I. Thus:

$$\frac{(A/U)_e}{(A/U)_m}=\frac{(\lambda_A A/\lambda_U U)_e}{(\lambda_A A/\lambda_U U)_m}=\frac{R_e}{R_m} \tag{8}$$

where R_e is the ratio of the number of alpha-particles emitted by actinouranium, $(\lambda_A A)_e$, to the number of alpha-particles emitted by the uranium I, $(\lambda_U U)_e$, in the terrestrial sample while R_m is the same "activity ratio" measured in the meteorite sample. Then equation 7 becomes:

$$(t_m-t_e)=\frac{ln(R_e/R_m)}{\lambda_A-\lambda_U} \tag{9}$$

Substituting the known values of the decay constants,[1]

$$\lambda_A = 0.98 \times 10^{-9}\ \text{years}^{-1}$$
$$\lambda_U = 0.153 \times 10^{-9}\ \text{years}^{-1}$$

[1] A. F. Kovarik and N. I. Adams, Jour. App. Phys., **12**, p. 296, 1941; A. O. Nier, Phys. Rev., **55**, pp. 150–153, 1939.

and converting to common logarithms through the relationship $\log x = 2.303\ ln\ x$, equation 9 becomes:

$$t_m - t_e = 2.8 \times 10^9 \log (R_e/R_m) \text{ years} \tag{10}$$

It will be noted that the coefficient of equation 10 is approximately equal to the age of the earth; thus the "activity ratio" R of actinouranium to uranium I has decreased by nearly a factor of 10 during the lifetime of the earth. Equation 10 also displays the fortunate fact that these differential age measurements depend on the ratio of two quantities which are to be determined experimentally, using the same apparatus and techniques on the two samples. Thus minor errors in measurement tend to cancel out, and the apparatus does not necessarily need to be calibrated on an absolute basis, since only ratios are wanted. Moreover, the final quantity sought, $(t_m - t_e)$, depends only logarithmetically on this measured ratio of the activity ratios R_e and R_m.

To derive the equations governing differential age measurements using the specific radioactivity of potassium, we return to equation 7 and write its analog for the case of the potassium isotopes. In any given sample of potassium, let:

N = number of atoms of the radioactive potassium isotope K^{40}
K = number of atoms of stable potassium
λ = total radioactive decay constant of K^{40}

Then remembering that the decay constant of a stable isotope is zero, we have by analogy with equation 7:

$$ln\ (N/K)_e/(N/K)_m = \lambda(t_m - t_e) \tag{11}$$

Again, as in equation 8, we can write:

$$\frac{(N/K)_e}{(N/K)_m} = \frac{(\lambda N/K)_e}{(\lambda N/K)_m} = \frac{S_e}{S_m} \tag{12}$$

where S_e and S_m are the specific radioactivities of potassium samples from terrestrial and meteoritic samples. Terrestrial potassium is[1] only 0.011 per cent K^{40}, while its specific radioactivity[2] S_e is about 23 beta rays per second per gram of potassium element. Probably K^{40} decays by orbital electron capture[3] as well as by negatron beta ray

[1] A. O. Nier, Phys. Rev., 48, p. 283, 1935.

[2] Mühlhoff, Ann. der Physik, 399, p. 205, 1930.

[3] R. E. Marshak, Phys. Rev., 61, p. 431, 1942.

emission. The fraction of the atoms decaying by orbital electron capture appears to be only 3 per cent, and would have the effect of increasing λ in equation 11 by 3 per cent. The decay constant of K^{40} is about 0.43×10^{-9} per year, corresponding to a half-period of 1,600 million years.

Substituting equation 12 and the numerical constants in equation 11 we have, for differential age measurements based on the specific radioactivity of potassium:

$$t_m - t_e = 5.3 \times 10^9 \log (S_e/S_m) \text{ years} \tag{13}$$

MEASUREMENTS RELATING TO THE URANIUM METHOD

To determine the differential age of the uranium atoms in two specimens, as indicated by equation 10, it is necessary to measure the ratio of the number of actinouranium atoms decaying to the number of uranium I atoms decaying per unit time in the sample of uranium. The relatively small difference between the range of the alpha rays from actinouranium and uranium I makes it impracticable to attempt to distinguish experimentally between the alpha rays from these two isotopes in any sample of uranium.[1] Therefore the measurement of the activity of actinouranium is made by separating from the sample one of its long-lived decay products, protactinium, with which it is in radioactive equilibrium. Similarly the activity of the uranium I isotope is determined by measuring the activity of one of its radioactive decay products, radon, with which it will be in radioactive equilibrium.

Experimental techniques of adequate sensitivity and accuracy are available for determining the radon content of rocks and meteorites.[2] The sample of rock or meteorite to be tested is boiled in a direct fusion vacuum furnace, operating at about 2000° C., thus releasing the inert radioactive gas, radon, from the sample. This radon is then conducted to an ionization chamber where its alpha-ray activity is compared with that due to a known amount of radon from a radium standard. Due to radioactive equilibrium in the original sample the number of radon alpha-rays observed per minute is the same as the number of alpha rays per minute produced by the uranium I in the rock sample.

The separation and measurement of protactinium is much more laborious. The chemical methods developed for separating pro-

[1] T. R. Wilkins and D. P. Crawford, Phys. Rev., 54, p. 316A, 1938.

[2] R. D. Evans, Rev. Sci. Inst., 6, p. 99, 1935.

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tactinium from rocks and stony meteorites, both of which are rich in silica, have been described.[1] About fifty grams of the siliceous material is heated in aqua regia and then hydrofluoric acid to remove the silica. After the addition of zirconyl chloride to the solution, the protactinium is co-precipitated with zirconium phosphate by the addition of phosphoric acid, leaving uranium, thorium, iron, and magnesium in solution but precipitating lead, barium, bismuth, and their isotopes or homologs among the radioactive decay products present. To assure complete separation of the uranium from the protactinium and zirconium phosphate, this precipitate should be dissolved in *HF* and reprecipitated as phosphate. After being washed with *HCl* the precipitate is ignited to zirconium pyrophosphate, ZrP_2O_7, and weighed. The zirconium pyrophosphate is then taken up in hydrofluoric acid and solutions of the nitrates of barium, lead, lanthanum, and bismuth (each carefully purified of radioactivity), equivalent to 4 mg. of each element, are added. At this stage the many radioactive elements other than protactinium are co-precipitated with their isotopes or chemical homologs of barium, lead, bismuth, and lanthanum, as insoluble sulphates and fluorides, leaving the protactinium and zirconium in solution. The co-precipitation process, using barium, lead, lanthanum, and bismuth, is repeated three times.

Overall control measurements, in which all reagents but no rock was used, show that the procedure, omitting a second precipitation of zirconium phosphate, removes substantially all (97 ± 3 per cent) of a small added amount of uranium, or thorium, or polonium. The recovery of protactinium is 90 ± 6 per cent.

Each 50-gram sample of stony meteorite or terrestrial rock used was made to yield slightly over 100 mg. of zirconium pyrophosphate. This was divided into carefully weighed individual samples of about 40 mg. each. Each sample was finely ground and deposited from an ethyl alcohol suspension, onto a silver disk 11 cm. in diameter and 0.5 mm. thick. The area of the final thin solid sample is 56.7 sq. cm. Thus the source has a thickness which is equivalent for the absorption of alpha rays to only about 4.5 mm. of air, and 46 per cent of all the protactinium alpha rays produced within the sample will emerge with a residual range greater than 5 mm. of air. This is equivalent to saying that the general equations[2] governing the internal absorp-

[1] R. D. Evans, J. L. Hastings, and W. C. Schumb, Field Mus. Nat. Hist., Geol. Ser., 7, p. 71, 1939; Jour. Amer. Chem. Soc., 61, p. 3451, 1939.

[2] G. D. Finney and R. D. Evans, Phys. Rev., 48, p. 503, 1935. Equation 3.

tion of alpha rays in thin sources show that the loss of countable alpha rays will be less than 8 per cent in samples of the area, weight, and composition given above. The source is placed in a parallel plate ionization chamber and the alpha-ray emission recorded by means of a vacuum tube electrometer amplifier and a photographic recording galvanometer (see fig. 31).

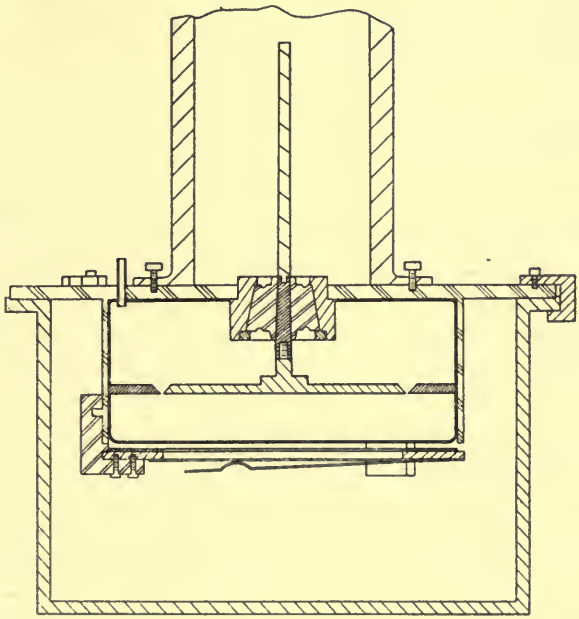

FIG. 31. Cross section showing construction of the parallel plate ionization chamber used for alpha-ray counting, which is similar to that developed by Finney and Evans.[1] The vacuum tube electrometer is located in an evacuated housing above the chamber. The collecting electrode is a circular brass disk, 10 cm. in diameter, and all inner parts are either made of spun electrolytic copper, or are silver plated, to minimize the background. The entire counting chamber is continuously flushed out with nitrogen which has been stored for at least one month, to cause the decay of any initial gaseous radioactive contaminants.

Parallel chemical manipulations and physical measurements were made throughout on a sample of the inside of a Pultusk stone, the outer skin being reserved, and on a terrestrial sample of granite from the Barryfield quarries, two miles east of Kingston, Ontario. Samples of the Pultusk meteorite were kindly furnished by Colonel Clifford C. Gregg, Director, and Mr. H. W. Nichols, Chief Curator, Department of Geology, Field Museum of Natural History, Chicago.

[1] Phys. Rev., 48, p. 503, 1935.

Table 1 summarizes the results obtained on these two materials.

TABLE 1.—SUMMARY OF MEASUREMENTS OF RADIOACTIVITY ON THE PULTUSK METEORITE, AND A TYPICAL TERRESTRIAL GRANITE FROM ONTARIO

	Pultusk Meteorite	Ontario Granite
Total α-ray activity of protactinium, in α per hour per g. of original material	0.10 ±0.03	1.4 ±0.3
Radium content in 10^{-12} g. Ra per g.	0.023±0.005	0.28 ±0.02
Radon α-ray activity in α per hour per g.	3.1 ±0.6	37. ±3.
Activity ratio, R, of protactinium/radon or actinouranium/uranium I	0.032±0.014	0.038±0.009

The total alpha-ray activity of protactinium is obtained by correcting the observed alpha-ray count for the internal absorption of alpha rays within the thin source, for the contamination of the zirconium pyrophosphate with an assumed 3 per cent of the uranium originally present in the sample, for the 90 per cent overall recovery of protactinium in the zirconium pyrophosphate, and for the small activity of the reagents as determined from control runs. The average counting rates were calculated from least squares averages of all observations taken, including alpha-ray counts of about forty hours on the Pultusk specimen, forty hours on the granite specimen, and several hundred hours of background, blank, and control runs. The average background of the alpha-ray counter under the conditions of measurement was 13.3 ± 0.3 alpha rays per hour.

The radium content of both specimens was measured by the radon method described briefly above, and the radon alpha-ray activity given in Table 1 is computed from these radium values by taking the Curie unit as 3.7×10^{10} disintegrations per second per gram of radium. Finally the activity ratio of actinouranium to uranium I, which is the same as that of protactinium to radon, is given in the last line of the table.

Within the probable error of measurement these activity ratios are the same in the Pultusk meteorite and in the terrestrial granite specimen. If we take the actual numerical values as given, then the quotient of the activity ratios R_e/R_m equals 1.2 ± 0.5. Substitution of this value in equation 10 gives

$$t_m - t_e = (0.2 \pm 0.5) \times 10^9 \text{ years} \qquad (14)$$

Thus these measurements indicate that there is definitely no large difference in age between the uranium atoms in the Pultusk meteor-

ite and in a terrestrial granite, and they strongly suggest that the difference between the ages of the atoms in these two samples of the solar system is zero.

It may be remarked that the ratio of the activity of actino-uranium to uranium I as measured in terrestrial uranium minerals lies somewhere between 0.40 and 0.46, with a strong preference being given to the higher value.[1]

It was found impossible to apply the uranium method to the Homestead meteorite, owing to the extremely small radioactive content of this meteorite.

POTASSIUM DIFFERENTIAL AGE MEASUREMENTS[2]

The meteoritic samples of potassium chloride were separated from the center portion of the specimens of the Pultusk meteorite. Samples weighing about 10 grams were disintegrated by treatment with 47 per cent hydrofluoric acid and the excess hydrogen fluoride removed. The large iron and magnesium content was precipitated by ammonium hydroxide and ammonium bicarbonate. The filtrate from this separation was evaporated to dryness in platinum and the excess ammonium salt carefully decomposed. The residue was then treated repeatedly with perchloric acid and alcohol until a constant weight of the final potassium perchlorate was obtained. The potassium perchlorate obtained from several of these separations was combined and treated once more with a perchloric acid and alcohol separation in order to insure a uniform final product, which was then decomposed in a quartz dish at 550° C., leaving potassium chloride. Great care was exercised in the choice and testing of reagents in order that none would be used that had come in contact with any terrestrial potassium during its preparation.

Merck C.P. reagent potassium chloride was used as a terrestrial comparison sample of potassium.

Spectroscopic analyses were made on both samples of potassium chloride used. The terrestrial sample had a purity greater than 99.8 per cent and contained less than 0.01 per cent rubidium. The meteoritic potassium chloride was over 99.2 per cent pure, the major impurity being calcium. The meteoritic potassium chloride contained 0.15 per cent rubidium. Rubidium is also naturally

[1] A. O. Nier, Phys. Rev., 55, p. 153, 1939.

[2] W. C. Schumb, R. D. Evans, and W. M. Leaders, Jour. Amer. Chem. Soc., 63, p. 1203, 1941.

radioactive, but with the purities indicated and under the conditions of measurement used the correction for rubidium activity is only about 2 per cent.

During the course of the work 93.04 grams of meteorite were used and from this material 0.415 gram of potassium chloride was recovered. This amounts to 0.28 per cent potassium oxide in the original

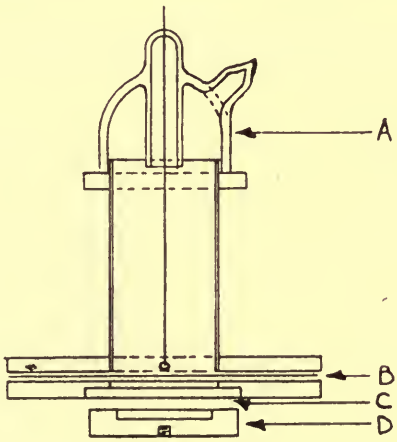

FIG. 32. "Bell" type Geiger-Müller counter and sample holder in half section. The counter is filled with dry air at a pressure of 60 mm. of mercury. All parts are of brass except A which is a glass top cemented to the tube. B is the mica window held in place with machine screws and picein cement. C is machined to receive the sample holder D and ensures uniform geometry to the sample and counter system. The window aperture in C is one inch in diameter.

meteorite, a value somewhat lower than that obtained by Rammelsberg in 1870 on another sample of Pultusk.

The measurements of radioactivity were made by placing the finely ground potassium chloride in a shallow cylindrical well (1.8 cm. in diameter) in a small brass disk (see fig. 32), the powdered sample being leveled by shaking and rocking. This sample was then brought close to a thin (21 microns) mica window of a bell-type Geiger-Müller counter. The amplifier for the counter was of the counting rate meter type, with photographic recording of the average counting rate by means of a drum camera and galvanometer.[1]

Nearly all the potassium chloride used in individual measurements can be recovered effectively and reused. In this manner 0.5 gram of potassium chloride could be used for many repeated

[1] R. D. Evans and R. L. Alder, Rev. Sci. Inst., **10**, p. 332, 1939.

measurements of the beta-ray activity. Owing to the very low specific radioactivity of potassium, reliable radioactivity measurements cannot be obtained on less than about 50 mg. of KCl. The area of the source disk is 2.54 sq. cm.; thus most of the samples had a thickness greater than 20 mg./sq. cm. Under these conditions the internal absorption of the soft beta rays of potassium is appreciable. In order to eliminate the effects of internal absorption, samples of varying total weight from 10 mg. to between 400 and 500 mg. were used. The net observed beta-ray counting rate per gram of potassium chloride is then plotted as a function of the amount of potassium chloride in the sample. Figure 33 shows the results on the terrestrial potassium, while figure 34 presents the observations on the potassium separated from the Pultusk meteorite. Extrapolation of these curves to a sample having zero weight, and hence zero internal absorption, gives the beta-ray activity per gram of potassium for the two specimens.

Comparison of figures 33 and 34 shows that the two curves have the same shape and the same intercept of 240 counts per minute per gram of potassium chloride. The probable error in each extrapolated intercept is about $\pm$ 10 counts per minute. Thus the ratio of the specific activities $S_e/S_m = 1.00 \pm 0.03$. Substitution of this ratio of the specific activities in equation 13 gives for the difference in the age of the atoms in Pultusk and in terrestrial material:

$$t_m - t_e = 0.00 \pm 0.06 \times 10^9 \text{ years} \qquad (15)$$

In agreement with the results obtained on uranium, but with a much smaller probable error, these measurements on potassium indicate that the age of the potassium atoms in the Pultusk meteorite is the same as the age of potassium atoms in the earth.

CONCLUSIONS

Comparative measurements have been carried out on the relative activity of the isotopes of uranium, and of the specific radioactivity of potassium, in terrestrial samples and in the Pultusk meteorite. These measurements show the same relative abundance of the isotopes of uranium and of potassium in both the terrestrial and meteoritic material. Hence they both confirm the basic assumption of the method in so far as the Pultusk meteorite is concerned, and show that the atoms of the Pultusk meteorite were created at the same time as the atoms in the earth.

To fix an absolute age for the time of formation of the Pultusk meteorite we would need to know the age of the atoms in the earth.

Measurements of radioactivity have been made by many methods on hundreds of terrestrial rocks and minerals in order to determine their "age," that is, the elapsed time since the rock or mineral was laid down in its present form and composition. These measurements show age values ranging continuously from zero for recent lavas up to about 2,000 million years. Age measurements by the helium method on rocks[1] and on magnetite minerals,[2] by the lead

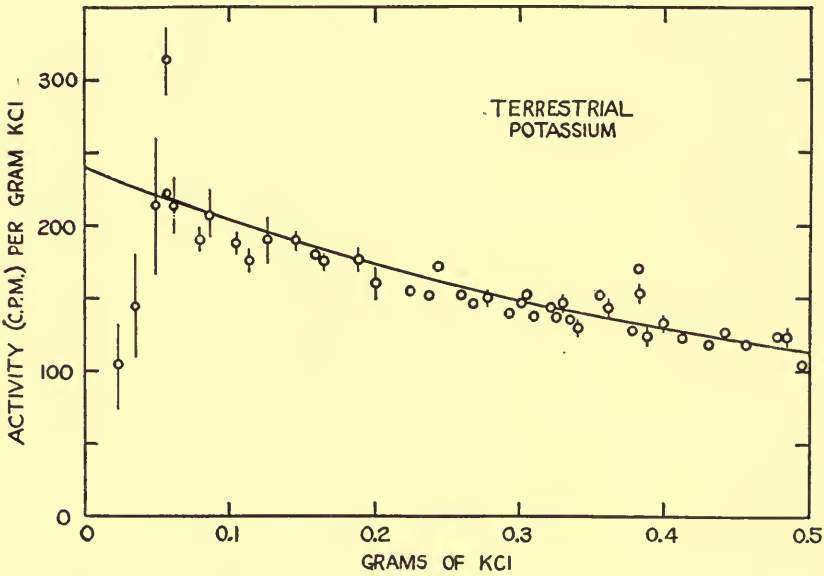

FIG. 33. Terrestrial potassium beta-ray activity per gram of KCl *vs* weight of sample used in the counter shown in figure 32. The probable error of each measurement is indicated by a vertical line through the experimental point. Extrapolation of this curve to a sample of zero weight eliminates the correction due to internal absorption within the various sources used.

method and by the lead isotope ratio method on thorium and uranium minerals[3] agree on the value of about 2,000 million years for the maximum age so far observed in any terrestrial occurrence. Considering the small fraction of the earth's crust which has actually been sampled it is entirely possible that one may eventually find

[1] For example: C. Goodman and R. D. Evans, Bull. Geol. Soc. Amer., **52**, pp. 491–544, 1941; C. Goodman, Jour. App. Phys., **13**, p. 276, 1942.

[2] P. M. Hurley and Clark Goodman, Bull. Geol. Soc. Amer., **52**, pp. 545–559, 1941.

[3] A. O. Nier, Phys. Rev., **55**, p. 153, 1939; A. O. Nier, R. W. Thompson, and B. F. Murphey, Phys. Rev., **60**, p. 112, 1941.

rocks or ores which are somewhat older than 2,000 million years, although present indications are that the distribution in the number of samples of high age decreases rapidly around 2,000 million years and may terminate near that figure.

It is generally agreed that an initially gaseous or molten earth would have solidified in less than 0.1 million years. But the span of time between the formation of an initially molten earth and the

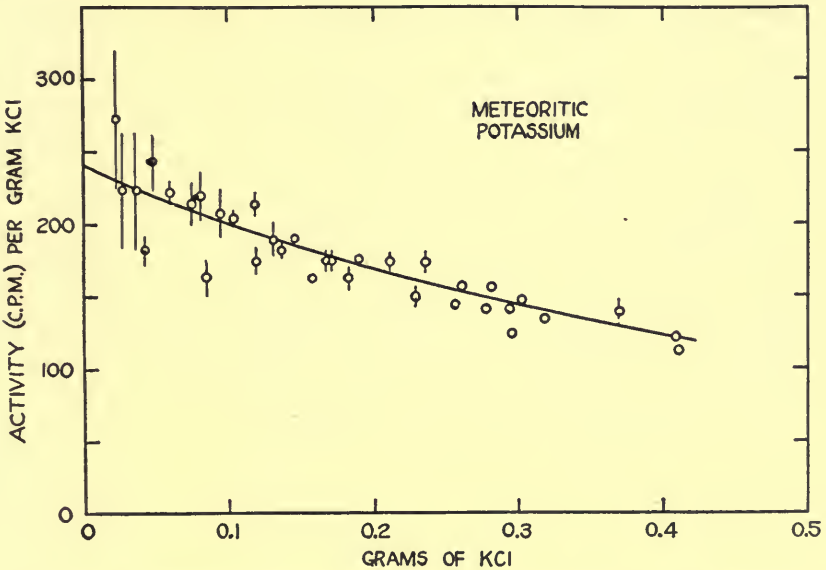

FIG. 34. Beta-ray radioactivity per gram of potassium chloride separated from the Pultusk meteorite, plotted in the same manner as figure 33.

creation of the atoms which it contains is entirely unknown. Present astronomical evidence favors the so-called short time scale of 10^9 to 10^{10} years. Thus, the stability of galactic clusters, the stability of wide binaires, the red-shift in extra-galactic objects, and other phenomena are compatible with a universe having essentially the same age as the earth.[1]

The isotopic abundance ratios have been determined with the mass spectrograph for all known elements. A general rule, apparently related to the fundamental laws of nuclear forces, is that isotopes of even atomic weight are always more abundant than isotopes

[1] Louderback, Evans, Gutenberg, Kuiper, Tolman, and Epstein, "Symposium on the Geologic and the Cosmic Age Scale." Science, 82, p. 51, 1935.

of odd atomic weight for elements whose atomic number is even. If this universal rule is extended to apply to the initial isotopic abundance of the uranium isotopes (even atomic number), then uranium I (atomic weight 238) should always have been more abundant than actinouranium (atomic weight 235). Equation 4 gives the isotopic abundance (A_o/U_o) at any time t_e years ago if A/U is the present[1] isotopic abundance. Substituting $(A/U) = 1/139$ and $(A_o/U_o) = 1$ in equation 4 we find that actinouranium and uranium I, if already created at that time, would have been equally abundant 6,000 million years ago, and at any earlier time the odd atomic weight isotope actinouranium would have been more abundant than uranium I, in contradiction to the general empirical laws governing the stable isotopes of all elements. This reasoning, if taken seriously, would require that the age of the atoms which compose the earth is something definitely less than 6,000 million years.

Within the relatively limited range of the sampled earth and the sampled meteorite population, all data agree on a period of 2,000 to at most 2,500 million years since the formation of the earth, and suggest the same order of magnitude for the age of the atoms of all elements so far studied in samples of the solar system.

[1] A. O. Nier, Phys. Rev., 55, pp. 150–153, 1939.

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